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EXCITED STATES OF CHLORINE NUCLEI

by

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A Dissertation Presented to the Faculty

of the

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Thesis
E53

Table of Contents

Introduction 1

1. Experimental Method 6

2. Experimental Results 44

Abstract

An investigation has been made of proton and alpha particle groups from deuteron bombardment, at various energies, of chlorine. Targets were HCl gas and evaporated AgCl layers, varying in isotopic constitution from 13 to 65 percent Cl^{37}.

Assignments of Q-values to isotopes were as follows: Cl^{35}: 6.27, 5.51, 5.09, 4.72, 4.30, 3.79, 3.42, 3.01, 2.71, 1.11, 0.78, and 0.07 Mev, corresponding to excitations in Cl^{36} of 0, 0.76, 1.16, 1.55, 1.97, 2.48, 2.85, 3.26, 3.56, 5.16, 5.49, and 6.20 Mev. Cl^{37}: 3.77, 2.55, 2.37, 1.86, 1.43, 0.29, and -0.38 Mev, corresponding to excitations in Cl^{38} of 0, 1.22, 1.40, 1.91, 2.34, 3.48, 4.15 Mev. The probable error for all Q-values is about 0.1 Mev; that for the excitations somewhat less.

A method of estimating corrections for peaking and other corrections required in the work is discussed.

Introduction

The existence of energy levels in atomic nuclei is one of the irritating problems in modern physics. The actual problem really no longer lies in the existence of such levels - after becoming accustomed to the discontinuous nature of energy processes in the relatively macroscopic field of atomic and molecular phenomena one rather expects it - the problem has become the lack of any adequate physical or mathematical representation of nuclear systems. It is certainly true that a great deal is known about the interactions of nuclei and about their energetic properties; it is equally certain that a great deal more is needed before a satisfactory theory of nuclei can be developed.

The methods available for the study of nuclear energy levels are several. One of the most prolific, and that used in this work, is the determination of energies of particles produced in nuclear reactions initiated by bombardment with energetic particles.

Pioneers in this activity with artificially accelerated particles were Cockcroft and Walton, who in 1930 with the now famous Cockcroft-Walton accelerator bombarded lead and beryllium targets, and in later work in 1932 observed the reaction $Li^7 (p,\alpha) He^4$ with protons of energy around 700 kev and energy release, or Q, of 14 Mev. It is of interest and amusing to note that this work involved also probably the first use of coincidence counting techniques. The measurements were accomplished by con-

tinuous recording on a tape of scintillations as seen by two observers watching screens on opposite sides of the target. A buzzer was kept sounding in the room so that the observers could not hear the clicks of each other's recording keys!

The results of Cockcroft and Walton were soon checked by Lawrence (L1) using the recently constructed 11-inch cyclotron at the Radiation Laboratory of the University of California (L2). At this time also Lawrence, Livingston, and White suggested the accessibility of nuclear energy levels through this kind of work.

Many investigations have followed. The kinetics of nuclear reactions have been worked out in great detail and have become commonplace; methods of analysis of data have become extremely refined. With improved methods has come the clear demonstration of the discrete nature of the energy changes in these reactions, and the observation of groups of product particles of differing and homogeneous energies shows very compellingly the presence of excited states in some of the nuclei involved. Numerous experiments have compared energy levels obtainable from different reactions (B1) and show in general that the group structure of protons from deuteron bombardment is due to the retention by residual nuclei of different but discrete and fixed amounts of the available energy, which amounts of energy are then liberated as gamma rays.

Many of the investigations of nuclear energy levels, and nearly all of those at this laboratory, have been made

on the (d,p) reactions in light nuclei. These reactions are studied by bombarding suitably prepared targets with deuterons of known energy, and measuring the numbers of protons emitted in various directions in discrete energy intervals at varying energies. (For a good 1937 treatment of the methods, see L3, for theory B2). Energy-momentum analysis involving the masses of the nuclei and particles (L3) shows the release of energy in definite amounts, independent of the bombarding energy, for a given target nucleus. The differences between these amounts, or Q values, have been shown to correspond to excited states (energy levels) in the residual nucleus. The numbers, energies, spacings, and relative probabilities of these levels, correlated with other properties displayed in other reactions of the nuclei of all the elements are expected ultimately to lead to a satisfactory nuclear theory.

As has been stated, much of the previous work has been on light elements. These are preferable from an experimental standpoint since their proton (hence energy level spectra are simple and the groups widely spaced. They are the more natural elements to attack from one theoretical standpoint as well in that presumably the simpler the system the simpler the theoretical treatment - this is conspicuously the case in the realm of atomic physics. This argument is not quite valid, however, as "exact" solutions can be consi-

ered to exist only for the lightest of nuclei; the theories which have been successful (in a qualitative way, which is as successful as such theories have been) in describing nuclear energy levels have been of a statistical nature, presuming large numbers of particles in the nucleus.

Recent trend has been toward investigations of heavier nuclei. The region of sulfur, chlorine, argon (A~35, Z~17) has been investigated with some care (D2, Z1, D3, etc.) - argon is still under study at this laboratory - but no work has been done on chlorine itself since that of Shrader and Pollard in 1940 (S1).

With this in mind and with an idea also of obtaining more information relevant to the increasingly important "magic numbers" (M4), (chlorine isotopes of mass 36 and 38, the residual nuclei of (d,p) reactions in chlorine, lie each side of the magic number 20 of neutrons) and investigation of excited states in the nuclei Cl^{36} and Cl^{38} has been undertaken.

Existing data are scanty. The original work of Shrader and Pollard reported six groups of protons and one of alpha particles from deuteron bombardment of silver chloride targets. The assignments and Q values were as follows:

Cl^{35} (d,p) Cl^{36}	Q = 6.31, 5.35, 1.50 Mev
Cl^{37} (d,p) Cl^{38}	Q = 4.02, 3.02, 2.10
Cl35 (d,α) S^{33}	Q = 9.1

These Q values indicate excited states in the residual nuclei of 1.0 and 1.92 Mev in Cl^{38} and of 0.96 and 4.81 Mev in Cl^{36}.

The only other measurement of excited states in chlorine nuclei are those of the Chalk River group (B6). These are (n,γ) measurements showing energies of 8.56, 7.77, 7.42 6.93, 6.62, 6.12, 5.72, 5.51, 5.01, 4.46, 4.06, and 3.62 Mev corresponding to excitations of 0.79, 1.14, 1.63, 1.94, 2.44, 2.84, 3.05, 3.55, 4.10, 4.50, and 4.94 Mev.
These have not yet been assigned to isotopes.

Mass assignments made by Shrader and Pollard were

Cl^{36}	35.9808
Cl^{38}	37.9806
S^{33}	32.9828

The present investigation was undertaken to attempt further to resolve the group structure whose presence was apparent, to make more accurate mass data available, and to essay some discussion of the excited state structure of the chlorine isotopes.

1. Experimental Method

After some consideration the decision was made to use a gaseous target. Such targets have yielded consistently good results in this laboratory (P1)(D2), seeming to give somewhat narrower groups than even very thin solid targets. A gas target of course cannot be surpassed for homogeneity under most conditions, and has the additional advantage of being flexibly and easily controlled in thickness. This latter advantage may be said to be an idealized one; the most practical procedure was found to be, once a target was prepared and got into the chamber, to leave it without tampering until the run was terminated.

The basic technique and most of the apparatus used have been described laboriously elsewhere. The bombardment chamber has been discussed by Davison (D2), and the counters and circuits used in conjunction with it were discussed by Martin (M4). Solid target observations were made using the bombardment chamber and method described by Martin (M4). Some of the work on both gaseous and solid targets was done with the large solid-angle counter described by Benson (B5). Counter fillings were argon at 15 and 20 centimeters of mercury pressure.

In the interest of reducing background and making the geometry of the system meaningful, the beam was defined by means of a square aperture 16mm on a side, set

eccentrically in the end of a 22 inch extension which was bolted to the cyclotron beam port with an O-ring seal. The aperture was placed eccentrically so as to pass the beam just inside the wall of the Davison chamber nearest the proton exit window, The whole chamber was insulated with a Teflon gasket and Teflon bushings beyond the beam aperture, and was used as collector from which the beam current was measured and integrated. The cyclotron magnet and deflector magnet currents were tuned generally to give maximum indicated beam current with this arrangement.

It was found that the gas target, HCl, was somewhat evanescent in nature while under bombardment: the target pressure might drop by fifty percent over a period of eight hours. This was at first believed to be caused by dissociation of the target and combination of the active chlorine with the chamber wall; the fact that the rate of disappearance of target decreased with time under bombardment seemed to substantiate this, as it indicated a saturation effect in the formation of a protective coating of chlorides (mostly zinc chloride) on the walls of the brass chamber. The same effect was later observed without bombardment, and from this it was concluded that the HCl was reacting directly with the chamber material despite the assurances of authorities (L5 and similar publications) that dry HCl hardly reacted with metals except aluminum. The aluminum foils used were never observed to be affected by the HCl except where exposed to the action of deuteron beam and

HCl together. In this case, though there was visible clouding of the aluminum surface, there was no detectable change in weight of the foils. This reaction could not then have been very extensive.

Because of the persistent and inconvenient loss of target, and because the volume of the Davison chamber and the associated manifold, tubing, etc. were excessive for satisfactory use of small quantities of target, a different approach was adopted in the later part of the work. A glass chamber was constructed and fitted with a small Bourdon-type vacuum gage (recalibrated against a mercury manometer to cm pressure instead of "inches of vacuum"), an evacuating connection, and a standard taper connection for filling. This chamber was sealed with de Khotinsky type cement (Cenco Sealstix) into a heavy brass flange machined to fit the base plate of the Davison chamber. The first glass chamber is shown set up and in use in Figure 1. The tubing, manifold, Selsyn-controlled foil changer, target bulb, and preamplifier are also visible in the photograph. This chamber was displaced slightly in the flange so that the beam would be centered in it. Its volume was about half that of the Davison chamber with the associated system required to get targets into it. The life of this chamber was short, as the deuteron beam soon honeycombed its unprotected end with fine cracks. A second glass chamber was constructed, of even smaller volume and this time with a disc of gold foil of about 0.002 inch thick-

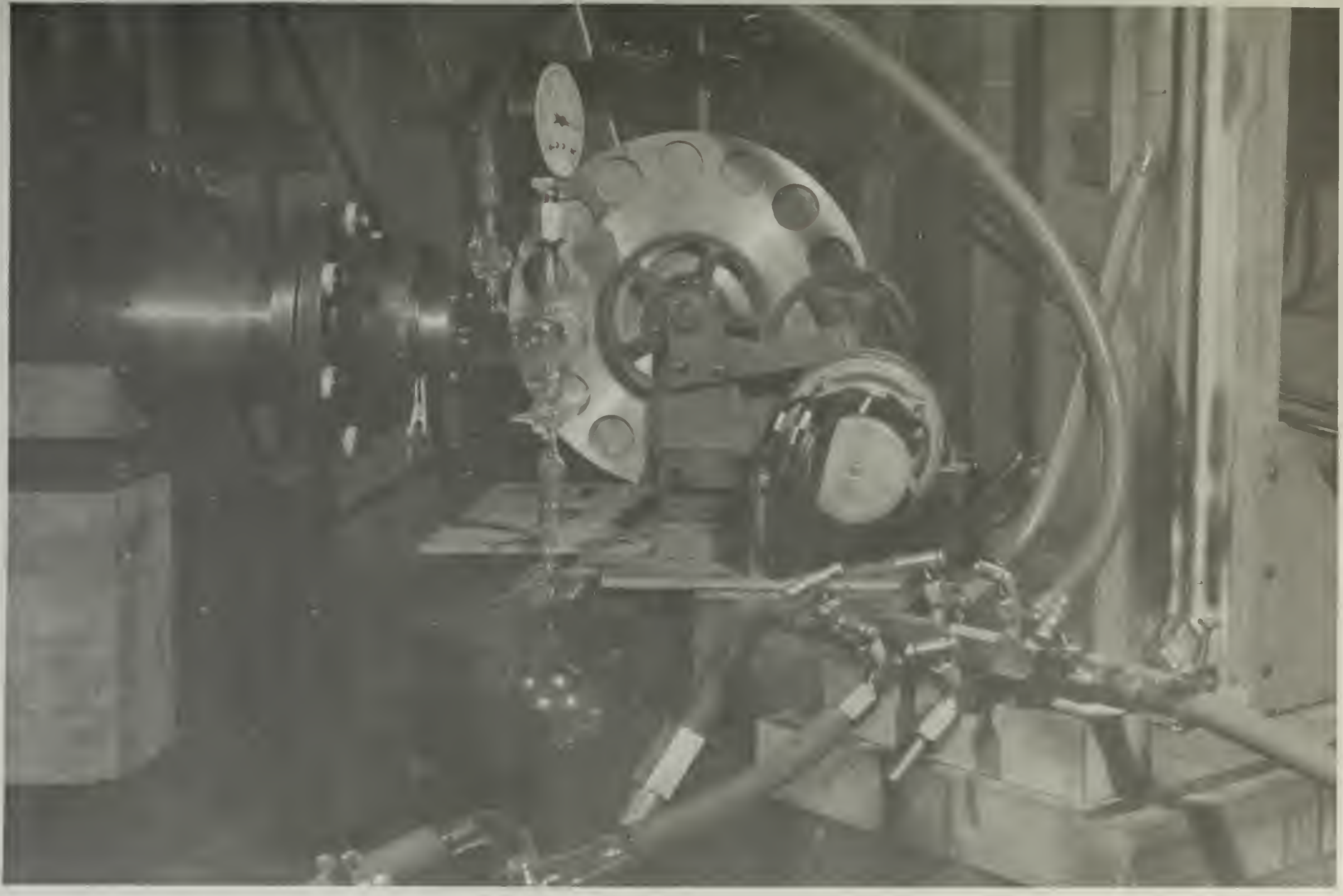


FIGURE 1

ness blocking the end of the tube beyond the proton port and the three connections. This foil was spot welded to a nickel wire, and the nickel wire to a wolfram wire which was brought out through a glass-to-metal seal. The foil was thus supported firmly and at the same time could be used as a beam collector to supplement the brass grid supporting the deuteron entrance foil.

Almost all of the beam energy measurements were made using the Davison chamber as a range cell with an end plate having a recess containing an insulated probe. The recess was isolated from the main chamber by an aluminum foil and was evacuated independently, being kept on the pump at all times when the chamber was in use. Other measurements of the beam energy were made with the range cell fitted to the solid target bombardment chamber in the arrangement shown by Sailor (S3).

The material chosen for target gas was HCl. This substance is available in a very high state of purity from the Matheson Co. of East Rutherford, New Jersey. The tank HCl is 99.5 percent pure with the principal impurities 0.2 percent acetylene and 0.3 percent chlorinated hydrocarbons. As HCl contains only one atom of chlorine per molecule it is well suited to isotopic separation by thermal diffusion. Its supposed nonreactivity is another argument in its favor.

Observations were made at 90° to the deuteron beam at all bombarding energies. It is not possible to use a gas target at zero degrees because of the prolific and complicated proton spectrum from aluminum (or from any other material such as mica which might be used in its place to isolatedthe target) which could not be separated from the protons from the target. Use of a gas target at any angle but 90° is in fact difficult and not recommended because of the excessive amount of stopping material in the gas itself in way of both protons and deuterons which results from the geometric requirement of not allowing any of the beam to strike any matter not part of the target which can be "seen" by the counter.

Target materials to enable assignment of excited levels to isotopes were available from two sources. The thermal diffusion isotope separation program at this laboratory, it was hoped, would be able to provide nearly pure HCl^{35} and HCl^{37} for study. From the Carbide and Carbon Chemicals Division of the Union Carbide and Carbon Corporation, Oak Ridge, Tenessee, can be obtained also silver chloride in small quantities enriched electromagnetically in chlorine content to 65.6% Cl^{37}, 34.4% Cl^{35}. 200 milligrams of chlorine in this form, corresponding to 126cc dry HCl at S.T.P., were obtained from this source with the intention of converting it to HCl for target purposes. The impurities in this material were determined spectroscopically to be:

Al	detected but less than	0.08%
Cu	" " " "	0.04%
Fe	" " " "	0.04%
K	" " " "	0.04%
Mg	" " " "	0.02%
Na	" " " "	0.08%
Si	____________	0.03%
Sn	detected but less than	0.03%

The separation of isotopes by thermal diffusion has been under investigation in this laboratory for some years. The phenomenon of thermal diffusion, i.e., gaseous diffusion as a result of a thermal gradient, was predicted independently in 1915 by S. Chapman (C4) and in 1917 by D. Enskog. Chapman soon pointed out the possibility of using the process for the separation of isotopes which could not be separated chemically, although his theory was not restricted to such a process, being equally applicable to any mixture of gases as long as there was a difference of molecular mass, size, or interaction among the kinds of molecules present. The theory was verified qualitatively experimentally by Dootson (D4). By 1934 Harmsen, Hertz, and Schütze had succeeded in separating Ne^{22} and H^2 to a high degree of purity, and in 1937 Clusius and Dickel hit upon the idea of cascading the thermal diffusion

into a continuous process by utilizing convective currents in a column in which a radial thermal gradient was maintained. Previous work had been restricted to hot-and-cold-bulb arrangments. With five 36 meter columns operating in series, Clusius and Dickel obtained 99.6% HCl^{35} and 99.4% HCl^{37}. No previous attempts to separate isotopes of chlorine had met any significant success, and this work was followed by a rash of theoretical and experimental investigations. Approximate treatments of the theory have been worked out by several authors (M2, J1, F1, D5, D8) and an exact solution has appeared recently for the differential equation of the diffusion column (M7). It is not proposed here to discuss the theory of thermal diffusion, which is most complex, but it is possible to describe the performance of the apparatus in simple terms which are set forth below, as taken from Jones and Furry (F1).

The transport equation for a column is written

$$\tau_1 = Hc_1c_2 - K\frac{dc_1}{dz}$$

where τ_1 is the transport of molecules of species 1 up the tube in grams per second, c_1 is the relative particle density of molecules of species 1 (the lighter one), $c_2 = 1-c_1$ is the relative density of the particles of species 2, and z is "a coordinate whose positive direction is up the tube". H and K are the transport coefficients, constants whose values depend upon the kind of molecules, the pressure, the mechanical characteristics of the

apparatus, and upon the temperature difference maintained across the column. K is the sum of the coefficients accounting for remixing: convection currents, ordinary diffusion, and parasitic currents induced by irregularities and asymmetries in the apparatus. As the conditions are symmetrical with respect to the heavier molecule, a transport equation for it

$$\tau_2 = Hc_1c_2 - K\frac{dc_2}{dz}$$

can be written with the positive direction of z now downward. The equation is generalized to apply to either component by letting c represent the concentration of the component being concentrated, ~~c~~ =1-c the concentration of the other component, the positive direction is that of increasing c, and τ is the transport rate of the material being concentrated. Then

$$\tau = Hc\bar{c} - K\frac{dc}{dz}$$

For discontinuous operation - that in which the system is allowed to reach equilibrium before any product is removed - the final state is characterized by the vanishing of τ all along the tube. With τ =0 the transport equation may be integrated to give

$$c/\bar{c} = e^{2A(z-z_0)}$$

where A= H/2K. Considering the ends of the column,

using subscripts f to designate the positive end and i to designate the negative end, one may invent a separation factor

$$q = \frac{(c/\bar{c})_f}{(c/\bar{c})_i}$$

whose equilibrium value is

$$q_e = e^{2AL}$$

where L is the length of the column. One may realize the equilibrium separation factor nearly fully by maintaining c_i at its natural or original value; this may be accomplished by continuously or intermittently replenishing the gas at the negative end of the column. Obviously it is desirable to make the quantity A as large as possible. It was believed possible as a result of recent work (S4,D6) to improve substantially upon the separation obtainable in HCl in a single column in a reasonable length of time by inducing a small controlled degree of "turbulence" in the column and by using an elevated cold wall temperature. The results were somewhat disappointing and are summarized in Table I for gas samples some of which were ultimately subjected to bombardment.

Table I

T_h	Tc	p	spacers	%Cl^{35}	q theor	q obs
700	15	72	7	82	-	1.8
700	70	72	7	81	-	1.8
850	84	74	7	86.2	3.75	2.1
900	81	72	132	87.5	3.75	2.4

Here T_h and T_c are the temperatures in degrees Centigrade of the hot wire and cold wall, respectively; p is the pressure in the column in centimeters of mercury; "spacers" refers to the number of small cross-pieces spotwelded to the hot wire (132 corresponding to one every inch); %Cl^{35} is the concentration of HCl^{35} in the upper end reservoir as determined by mass spectrometer; and q_{theor} and q_{obs} are the separation factors as estimated from theoretical considerations and as actually observed.

The best concentration of Cl^{37} attained was 31.4%, corresponding to a separation factor of about 1.4.

The column used in these efforts was 11 feet long and 3/8 inch in inside diameter; the hot wire was of 15 mil platinum.

The labors involved and the figures cited above were provided by Mr. A.Z. Kranz.

The method of handling the small quantities of HCl used was taken from Sanderson (S5) and consisted essentially in

1. Evacuating the container where the HCl was desired to be

2. Opening the connecting cock or cocks between the reservoir and receiver

3. Reducing the temperature of the receiver with liquid air so that the HCl would freeze on its walls

4. Closing the connection.

Liquid air temperature was found to be a very effective HCl pump, and the method also permitted purifying the gas of the universal contaminants nitrogen and oxygen simply by pumping off the residual gas while the HCl was frozen. In this way small but quite pure samples of HCl (perhaps 50cc at $\frac{1}{2}$ atmosphere) could be obtained.

In view of the evanescent quality of gas targets mentioned above, it was decided to use the small quantity of enriched silver chloride obtained from Oak Ridge in the form of solid targets. These were prepared by evaporation in vacuum on thin gold leaf. A tantalum ribbon whose center was shaped into a cup about 2mm square and 2mm deep was used as a filament and holder for the material to be evaporated, and was bent so that the cup was located well up inside a tubular glass baffle whose constricted upper end defined a circular area of deposition upon the gold leaf. The glass baffle served to intercept all the AgCl which did not go to the target and at the same time permitted visual observation of the progress of the

evaporation from the filament. This method proved satisfactory only for the production of very thin targets, as at high vacua silver chloride boils violently at temperatures little above the melting point and deposits obtained by using large (50mg) quantities of the material or by moving the cup nearer the target were invariably granular. This seemingly independent of the filament temperature used. It was never found possible to make a target thick enough to permit detection of the highest energy groups of Cl^{35}, either with enriched AgCl or the natural material. It was thought that AgCl should make a nearly ideal target - before it was appreciated that the effect of the deuteron beam upon it would be just the same as that of white light but much intensified, leaving fine black deposits of metallic silver upon the gold backing.

The procedure followed in obtaining data was uniform and conventional. Considerable difficulty was experienced in early stages of the work, particularly with the low yield high energy groups, in obtaining significant yields without excessive backgrounds and large target thicknesses with resulting high group widths and general bad resolution. Defining the beam by means of the 16mm square iris (replacing a 32mm round one) reduced background difficulty considerably in addition to reducing the energy width of the beam somewhat. It was not until the small volume glass chamber was used

that the background became consistent and reasonably small, allowing reliable integral curves to be taken on the end group. It was contrived to take the equivalent of a total of about ten complete runs over the whole range of proton energy accessible. The search for additional yield was carried out to 150 and 200 centimeters on several occasions without finding anything except the high Q group from nitrogen.

It was found possible to take integral curves on several of the inner groups using the evaporated AgCl targets, because the yield of the outer groups was so small. The relative yields obtained from the solid targets must be regarded as suspect, however, except for adjacent groups, because of the decomposition of these targets by the beam.

Before filling a gas chamber with HCl it was always dried thoroughly by flushing about a half-dozen times with air dried in a liquid air trap. This was a time-consuming procedure, but it was found to be necessary as the aluminum foils were subject to attack if it was not followed. As has been mentioned, the brass was attacked anyway, but the rate of absorption of the HCl by the chamber decreased with time - presumably with the formation of a protective coating.

A condition contributing to uncertainties in the values of Q's derived from this apparatus is the lack of regulation other than "intermittent manual" type of

the deflector magnet which bends the external beam clear of the main magnet yoke. The exciting current must be adjusted frequently; as a consequence errors from this source cannot be eliminated entirely. The dependence of the mean beam energy upon this current is not great, however (80 kev between 94 and 100 amperes), so the net effect is estimated as a small increase in the probable error.

The stopping power of chlorine was determined by making one beam energy determination with HCl in the range cell instead of air. This gives a direct conversion from centimeters of HCl to centimeters of air for deuterons averaged over the range from zero to the beam energy. This value was used in all calculations, as the relative magnitude of the energy change from slowing of the beam by the gas before it reaches the target region is much greater than that imposed by the small amount of gas in way of the emitted protons. The equivalence found was that 1 cm HCl = 1.12 cm air, yielding an atomic stopping power for chlorine of 2.04 if combined with an average value for the stopping power of H (L3).

In the experimental work it has not always been possible to resolve clearly the closely spaced groups. It is possible to derive extrapolated ranges from such ill-resolved groups by the method of "construction" which

[illegible]

[illegible]

[illegible]

has been employed by many investigators, but the results are not to be believed as readily as those obtained from well-defined, isolated groups. The same considerations apply to relative yields, which can be caused to vary widely by small variations in width used in the construction. Figure 8 shows the resolution into groups of one of the runs on solid silver chloride by this method. The procedure must be used with discretion; it is after all not difficult to "resolve" into groups of any width desired a horizontal straight line.

The ranges measured were converted to energies using the Cornell 1937 range-energy relation. The Q-values were derived from the non-relativistic energy-momentum relation for 90° observation

$$Q = \frac{M_P + M_R}{M_R} E_P - \frac{M_R - M_I}{M_R} E_I$$

where subscripts I, R, and P refer to incident, residual, and product particles, respectively. Subscript D (for deuteron) appears in the section following on corrections; no other incident particles were employed in this work.

It has here long been the practice to calculate Q-values from the "extrapolated" energy of particle groups - the value obtained by linear extrapolation of

the steepest slope of the numbers-energy or numbers-range curve to the baseline - and the similarly "extrapolated" energy of the integral curve of the deuteron beam. This does not lead to very large errors ($>$ 150 kev) in general, but it does lead to errors which can be corrected to become smaller than other and unavoidable errors. Motz in 1949 (M1, M2) showed how such errors could be corrected quite precisely on the assumption that the distribution in energy of the beam was representable by a Gauss error function. The present work, without discounting Motz' work but discounting the assumption, derives reasonable corrections in a simple way by an extension of Motz' treatment. It is advisable, however, to begin at the beginning with a treatment of the Gauss distribution and its characteristics.

Gauss distribution

Mean value x_o, Parameter α, Total number N.

$$dn = \frac{N}{\sqrt{\pi}}\, e^{-\left(\frac{x-x_o}{\alpha}\right)^2} dx = c\alpha e^{-z^2} dz \quad \text{with } z = \frac{x-x_o}{\alpha}$$

$$\frac{dn}{dz} = y = c\alpha e^{-z^2}, \quad \frac{dy}{dz} = -2c\alpha z e^{-z^2} = -2zy$$

$$\frac{d^2y}{dz^2} = -2\left[1-2z^2\right]y$$

whence $z = 1/\sqrt{2}$ at steepest slope.

$$x = x_o + \alpha/\sqrt{2}$$

Here $y = c\alpha e^{-1/2}$, $dy/dz = -\sqrt{2}\, c\alpha e^{-1/2}$ The extrapolated intercept $z_{ext} = 1/\sqrt{2} + \dfrac{c\alpha e^{-1/2}}{\sqrt{2}\, c\alpha e^{-1/2}} = \sqrt{2}$

$$x_{ext} = x_o + \alpha\sqrt{2}$$

Integral curve

$$n = c\alpha \int_z^{\infty} e^{-z^2} dz = \frac{N}{\sqrt{\pi}} \int_z^{\infty} e^{-z^2} dz$$

$$\frac{dn}{dz} = c\alpha e^{-z^2} = y\, ; \quad \frac{d^2n}{dz^2} = \frac{dy}{dz} = -2zy = 0$$

at point of steepest slope, whence $z = 0$, $x = x_o$ at this point. Here also

$$n = N/2, \quad dn/dz = N/\sqrt{\pi}.$$

The extrapolated intercept then is

$$z_{ext} = \frac{N}{2} \Big/ \frac{N}{\sqrt{\pi}} = \frac{1}{2}\sqrt{\pi}$$

$$x_{ext} = x_o + \frac{\alpha\sqrt{\pi}}{2}$$

The difference then between the extrapolated intercepts of a Gauss distribution and its corresponding integral curve is

$$\left(\sqrt{2} - \frac{\sqrt{\pi}}{2}\right)\alpha = 0.528\,\alpha = 0.635\,\Gamma$$

α is the half width of the distribution at 1/e = 0.368 maximum height. At half-maximum the half width Γ of the distribution is given by

$$e^{-\left(\frac{\Gamma}{\alpha}\right)^2} = \frac{1}{2}$$

whence

$$\Gamma = \alpha\sqrt{\ln 2} = 0.832\,\alpha$$

A counter-absorption measuring system counts particles in a finite interval of range (energy), hence can only approach a genuine differential curve in its results. The counting interval depends upon the geometry of the counter itself and upon the circuits associated with it - "peaking", bias, etc. What the system counts is particles of range beyond that determined by the controlled amount of absorbing material in their path; the peaking of the circuits determines how far beyond. For a given amount of absorbing material, corresponding to a range (or energy) x, (or z),

$$n = c\alpha\int_{z}^{z+\Delta z} e^{-z^2}\,dz$$

For infinite counting interval (Δz; this amounts to

[illegible]

$$[illegible]$$

[illegible]

$$[illegible]$$

[illegible]

$$[illegible]$$

[illegible]

$$[illegible]$$

[illegible]

so biassing the counting circuits that all particles passing through the counter are counted) this is simply the integral curve analyzed above. As the counting interval is shortened, the right profile of the numbers-range curve remains unaltered until $\Delta x \sim 4\alpha$. The ordinate n of the number range curve is given by

$$n = c\alpha \int_{z}^{z+\Delta z} e^{-z^2} dz$$

$$= c\alpha \int_{0}^{z+\Delta z} e^{-z^2} dz - c\alpha \int_{0}^{z} e^{-z^2} dz$$

$$= N/2 \left[\Phi(z+\Delta z) - \Phi(z) \right]$$

where, again

$$z = \frac{x - x_0}{\alpha}$$

The integrals are now the well known (and tabulated) probability integral.

The minimum group width attainable with such an arrangement is limited by α. For a true differential curve ($\Delta z \to 0$) the half width at 1/e of the maximum is α; at ½ maximum it is 0.832 α as determined above. In this case, however, the yield (maximum ordinate) is zero. On the other hand the integral curve ($\Delta z \to \infty$) gives maximum yield (N) but infinite width. An optimum degree of peaking evidently lies somewhere between these extremes. If one accepts the ratio of maximum ordinate to half

width as a criterion of merit one may calculate graphically the optimum Δz.

The maximum ordinate of the curve occurs at $x = x_0 - \frac{1}{2}\Delta x$ or $z = -\frac{1}{2}\Delta z$. Its value is

$$n_{max} = c\int_{x_0-\frac{1}{2}\Delta x}^{x_0+\frac{1}{2}\Delta x} e^{-\left(\frac{x-x_0}{\alpha}\right)^2} dx = 2c\alpha\int_0^{\frac{\Delta z}{2}} e^{-z^2}\,dz$$

The group half width at $\frac{1}{2}$ maximum is given by $x-x_0+\frac{1}{2}\Delta x$ for such value of x that

$$c\int_{x}^{x+\Delta x} e^{-\left(\frac{x-x_0}{\alpha}\right)^2} dx = c\alpha\int_{z}^{z+\Delta z} e^{-z^2}\,dz = c\alpha\int_0^{\frac{\Delta z}{2}} e^{-z^2}\,dz$$

For $\Delta z = 0$ (true differential curve) the half width at half- maximum is $\sqrt{\ln 2} = 0.832$ ·

For $\Delta z = 1$, for example

$$\left[\Phi(z+1)-\Phi(z)\right] = \Phi\left(\tfrac{1}{2}\right) = 0.5205$$

determines

$$z = 0.404\,,\ \Gamma = 0.904$$

These are plotted on Fig. 2, with the maximum ordinates and peaking factors P. It is seen that a broad maximum in the peaking factor appears at $\Delta z = 1.9$; this serves as an approximate criterion of optimum peaking for best resolution of groups with low yield.

[illegible]

$$[illegible]$$

[illegible]

$$[illegible]$$

[illegible]

$$[illegible]$$

[illegible]

$$[illegible]$$

[illegible]

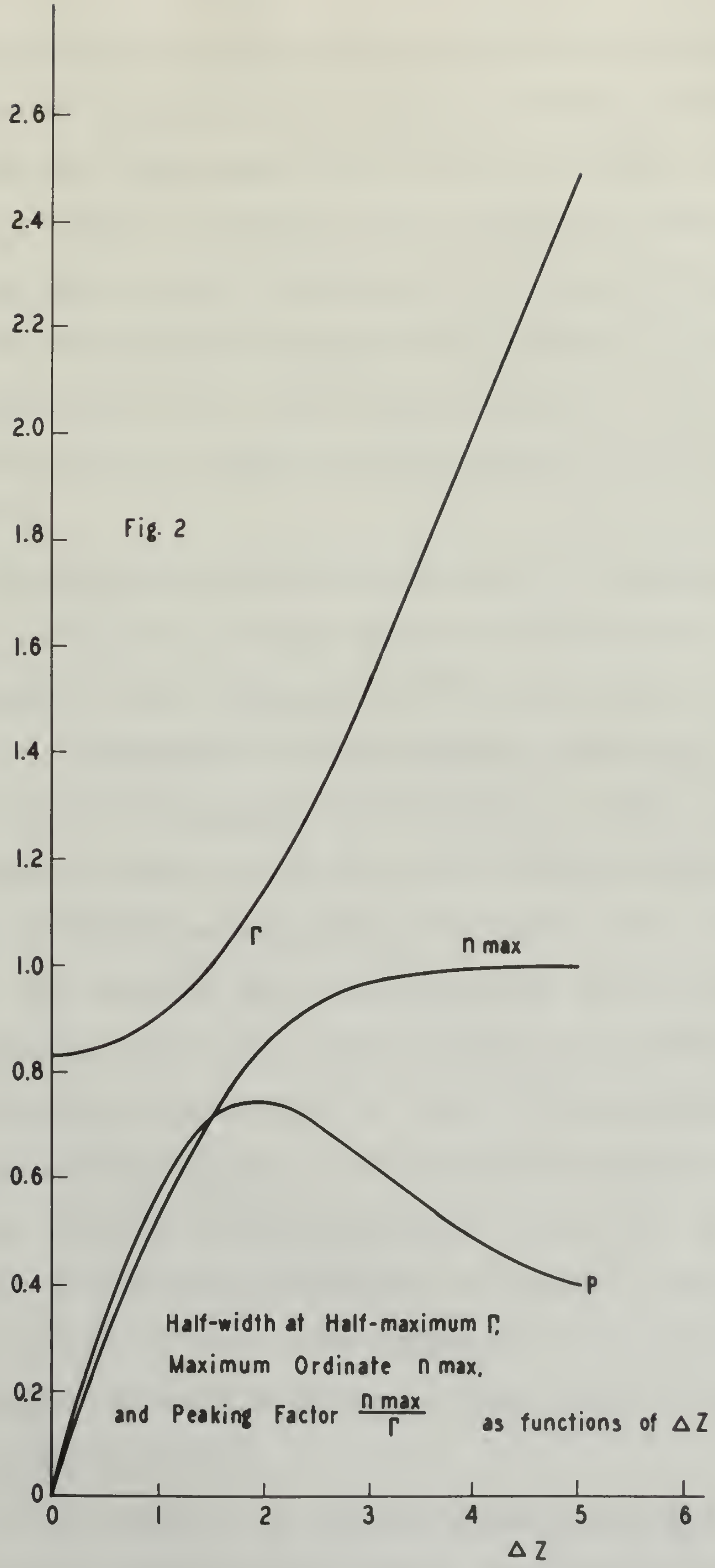


Fig. 2

Half-width at Half-maximum Γ,
Maximum Ordinate n max,
and Peaking Factor $\frac{n_{max}}{\Gamma}$ as functions of Δ Z

For very prolific groups one may accept the decreased yield to be expected from increased peaking in order to obtain the improvement in resolution (which is, however, slight) resulting from diminished group width. This may work to the advantage of the experimenter in that the most prolific groups are generally those of low energy which are most closely spaced. (Corresponding to transitions to highly excited states in the residual nucleus.)

Extrapolated ranges (or energies) of distributions of this sort will be very near the extrapolated range of the integral curve ($z_{ext} = \frac{1}{2}\sqrt{\pi}$) for large Δz; as peaking is sharpened the extrapolated range will approach that of the differential distribution. (Also, as has been shown by Motz and above, the abcissa of the maximum of the distribution approaches the mean value for the groups. The reasons for not using this value will appear.) Extrapolated ranges are shown on Fig. 3 in terms both of the distribution parameter α and of the position of the extrapolated range in the interval between the two limiting values. The extrapolated ranges may be calculated from the distribution equations or measured from plots of the distributions. Both figures 2 and 3 have been calculated from tabulated values of the exponential and probability functions.

The calculation of the extrapolated ranges is rather tedious but does show that Motz' graphically determined

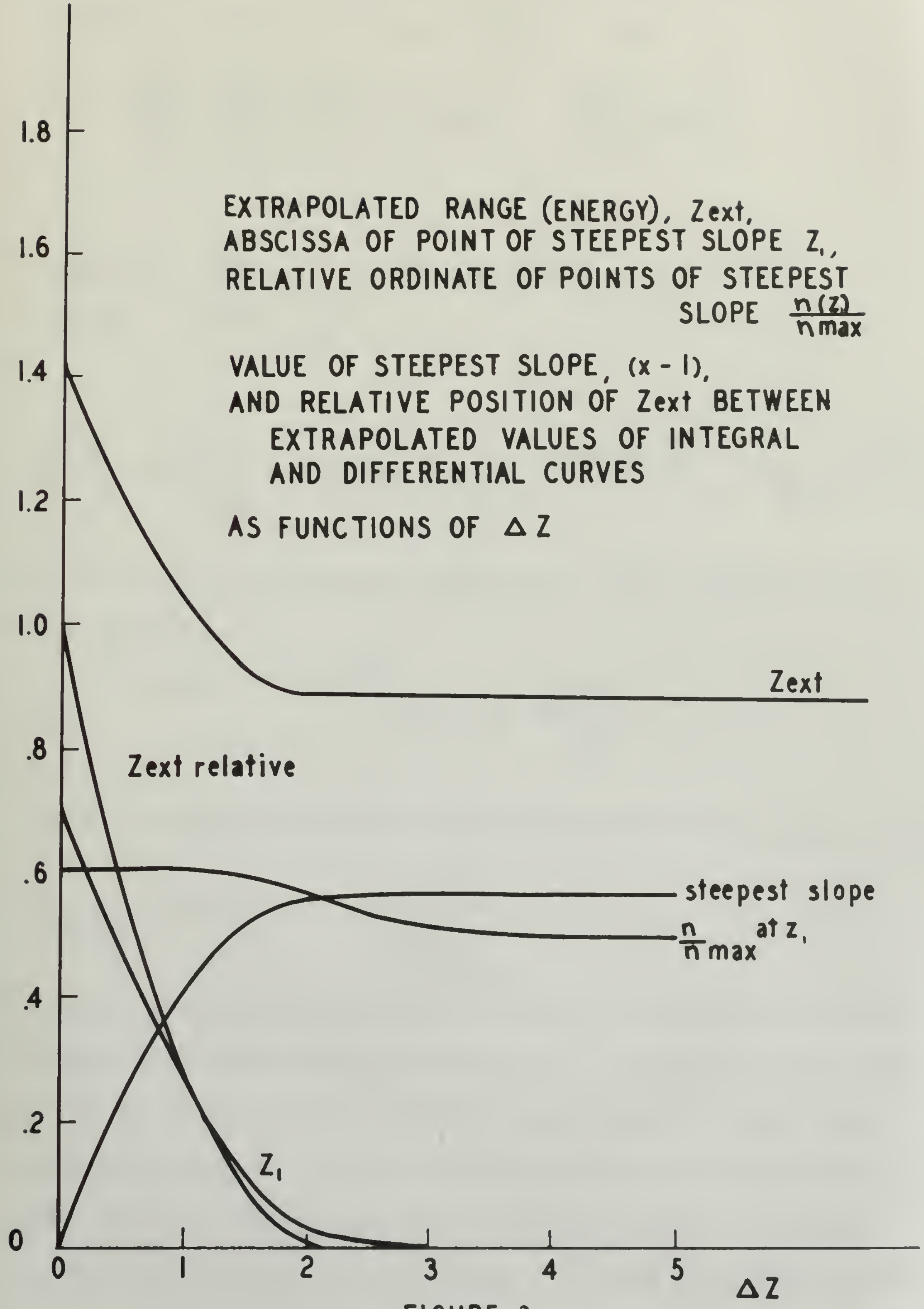

EXTRAPOLATED RANGE (ENERGY), Zext,
ABSCISSA OF POINT OF STEEPEST SLOPE Z₁,
RELATIVE ORDINATE OF POINTS OF STEEPEST SLOPE $\frac{n(z_1)}{n\,max}$
VALUE OF STEEPEST SLOPE, (x - 1),
AND RELATIVE POSITION OF Zext BETWEEN EXTRAPOLATED VALUES OF INTEGRAL AND DIFFERENTIAL CURVES
AS FUNCTIONS OF ΔZ
Zext
Zext relative
steepest slope
$\frac{n}{n\,max}$ at z_1
Z_1
1.8
1.6
1.4
1.2
1.0
.8
.6
.4
.2
0
0
1
2
3
4
5
ΔZ

FIGURE 3

values are too high by small amounts in the region between the extreme values of Δz. One has

$$n = \frac{N}{2}\left\{\Phi(z+\Delta z) - \Phi(z)\right\}$$

$$\frac{dn}{dz} = \frac{N}{\sqrt{\pi}}\left\{e^{-(z+\Delta z)^2} - e^{-z^2}\right\}$$

$$\frac{d^2n}{dz^2} = \frac{N}{\sqrt{\pi}}\left\{-2(z+\Delta z)e^{-(z+\Delta z)^2} + 2z\,e^{-z^2}\right\} = 0$$

at the point of steepest slope z_1. This equation can be simplified to

$$e^{2z_1\Delta z + (\Delta z)^2} = 1 + \frac{\Delta z}{z_1}$$

which is solved from the tables for any value of Δz. Then

$$z_{ext} = z_1 + \frac{n(z_1)}{\left(\frac{dn}{dz}\right)_{z_1}}$$

The ordinates of the points z_1 and the slopes at these points are also plotted on Fig. 3. It is seen that the point of steepest slope falls very near 0.6 times the maximum ordinate for all the most important values of Δz. This fact is useful in extrapolating ranges and could serve as an index of mean range. If one can predict the

z coordinate and percent maximum ordinate of the point of steepest slope as a function of $\triangle z$ it is obvious nonsense to use the extrapolated value at all. If the position of the maximum is uncertain this should be a good method.

This analysis gives a very good method for treating data initially representable by a Gauss distribution. It has been made as detailed as it has because the Gauss distribution represents a distinct and important entity in this kind of study - it being the form of the distribution generated by statistical processes such as straggling.

One must now take into account the fact, readily observed, that the shape of the curves taken with the equipment used in this work is not Gausssian and indeed cannot be.

A study of the energy distribution of the cyclotron beam has been made using a method slightly more refined than those employed by previous investigators. In this study the usual arrangement involving a range (gas absorption) cell was used but with the added feature of two sensitive galvanometers so arranged that one indicated at all times the total beam current while the other indicated the current getting through the range cell. In this way an accurately controlled plot of percent transmission vs. absorption could be made and, ideally, repeated. The measurements made yielded, however,

some rather surprising results:

1. Strong evidence that the energy distribution of the beam is formed of a composite of at least two distributions of different mean energies.

2. The relative intensities of the two principal peaks are subject to fluctuations.

3. The separation of the two principal peaks is subject to fluctuation.

(Selected curves shown on Figs. 4A and 4B.) The actual cyclotron beam distribution may be said to be "closely Gaussian" about halfway up the profile of the integral curve,i.e., between the mean and upper limit energies. At lower energies pronounced deviations appear resulting in distortions of the upper part of the differential distribution whose ultimate result is that the location of the maximum (if there is only one) is not well defined. It is considered likely by the writer that the dual peak effect is caused by scrambling in the cyclotron of aggregates of deuterons which have enjoyed total numbers of accelerations differing by one or two. Such effects could be contributed by orbit instabilities in the cyclotron itself, variations in the main magnet current (relative current readings in the beam measurements were observed to be very sensitive to fluctuations in magnet current), and the fact that the total number of accelerations imposed upon the particles in this cyclotron is unusually large. This number is estimated to be 50 for deuterons

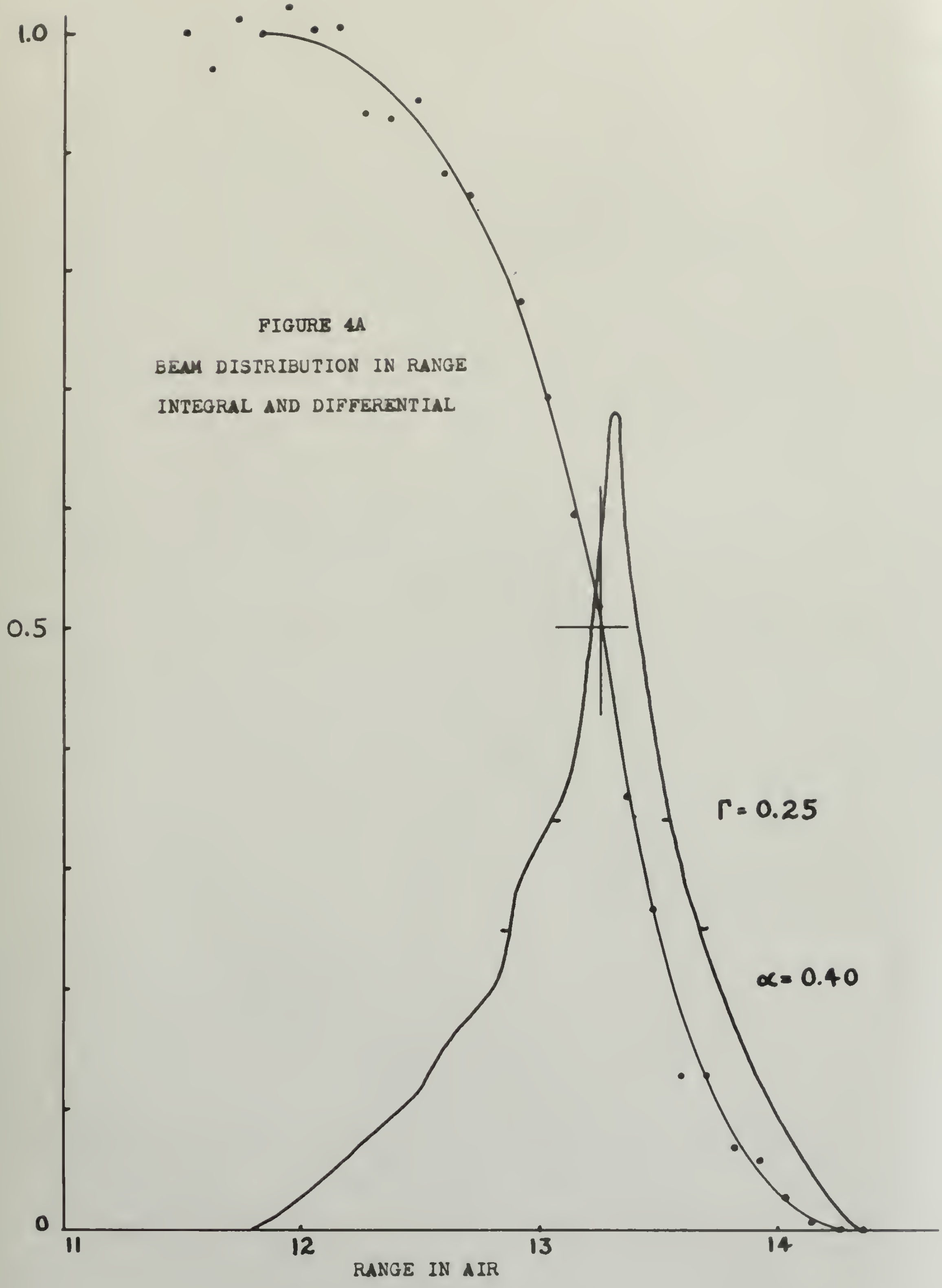


FIGURE 4A

BEAM DISTRIBUTION IN RANGE

INTEGRAL AND DIFFERENTIAL

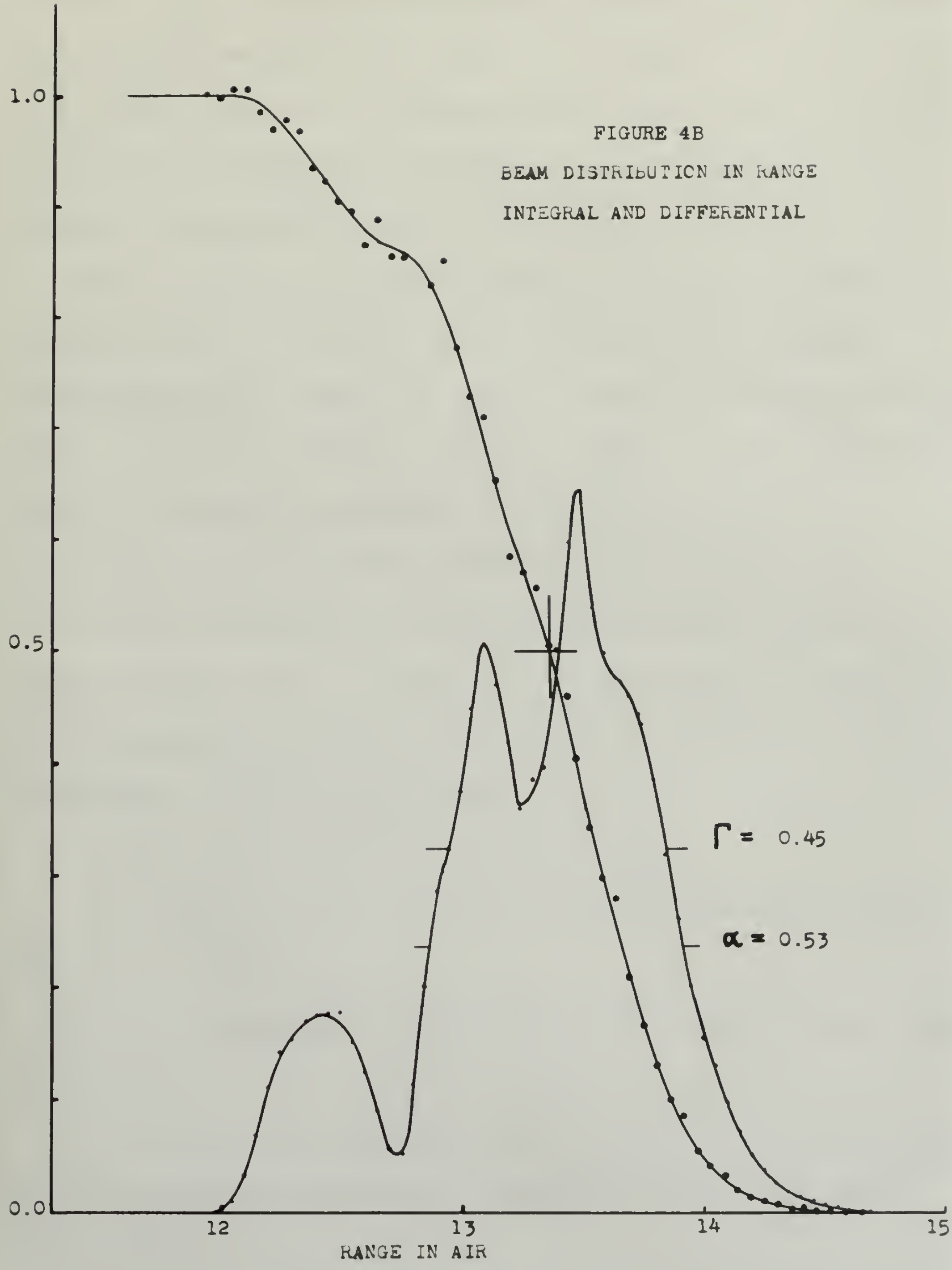


FIGURE 4B

BEAM DISTRIBUTION IN RANGE

INTEGRAL AND DIFFERENTIAL

of 4 Mev; the estimated energy increment of 70 kev per acceleration (with 50° phase lag) (R1) is not incompatible with this hypothesis and the observed beam width. It is easily appreciated that the result of adding parts of a small number of distributions (even though probably Gaussian) of different energies is not likely to be an overall distribution which is Gaussian in contour. It is quite possible, however, for such an overall distribution to have a profile very like that of a Gauss distribution and still to have a broad uncertainly located maximum or even multiple maxima. This is the condition which is believed to exist and to account for the anomalously wide energy spread of the external beam of the Yale cyclotron. A study of published measurements of this beam, as well as of the shape of isolated groups measured with it, offers strong support for this hypothesis. (P1, P2, M1, M2.)

Given this set of conditions, i.e.,

1. Approximately Gaussian shape of the high energy edge of the beam

2. Irresponsible behaviour of the rest of the beam distribution

3. Large probable error in measured energy of beam, the following conclusions regarding analysis of data were drawn:

1. Extrapolated range would be more reliable and

reproducible than range of group maximum as an index of mean range.

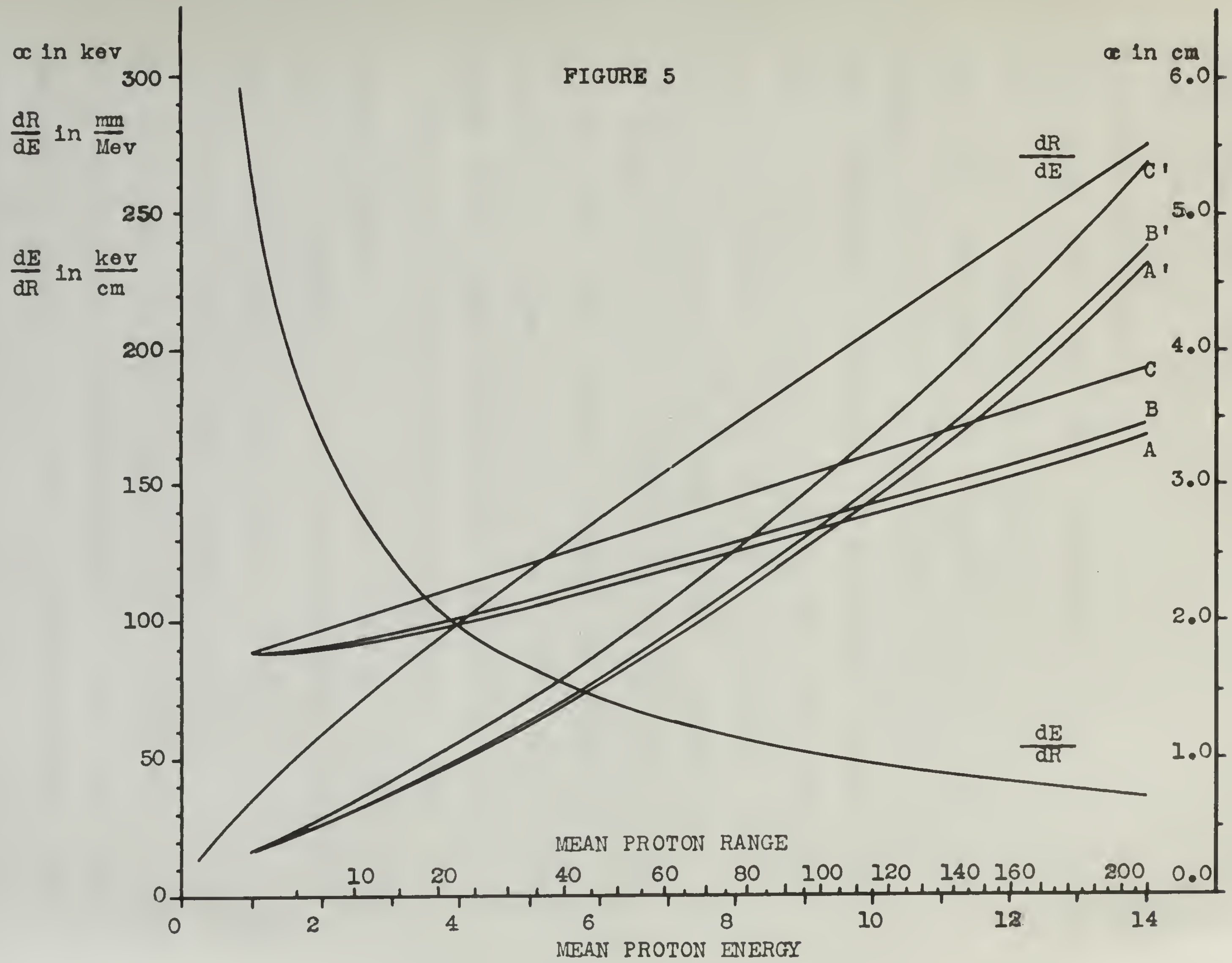
2. Use of "extrapolated energy" (converted from extrapolated range) and extrapolated beam energy from either integral or differential curves to determine Q-values would with suitable corrections yield all the accuracy of which the system is capable.

3. A combination of mathematical analysis and hammer-and-crowbar techniques should allow calculation of "suitable corrections" as functions of energy and of difference between extrapolated approximation and accurately established Q-value for such accurate values as may be available.

To permit analysis on this basis the curves of Fig. 6 were calculated, using the curves of Fig.5. Rigorously the analysis still applies only to results obtained with a beam whose energy distribution is "Gaussian", but it is believed that errors introduced by assuming the beam energy shape to be Gaussian are smaller than other errors introduced by energy fluctuations, errors of measurement, etc.

Figure 5 shows plots of dR/dE and dE/dR versus energy for protons, calculated from the "range exponent" given by Livingston and Bethe (L3). Also shown on Fig. 5 are values of predicted group parameter α_p calculated by compounding the three effective parameters of beam,

FIGURE 5


α in kev
$\frac{dR}{dE}$ in $\frac{mm}{Mev}$
$\frac{dE}{dR}$ in $\frac{kev}{cm}$
300
250
200
150
100
50
0
α in cm
6.0
5.0
4.0
3.0
2.0
1.0
0.0
$\frac{dR}{dE}$
C'
B'
A'
C
B
A
$\frac{dE}{dR}$
MEAN PROTON RANGE
10
20
40
60
80
100
120
140
160
200
0
2
4
6
8
10
12
14
MEAN PROTON ENERGY

range straggling, and angle straggling by the formula $\alpha_p^2 = \alpha_b^2 - \alpha_s^2 - \alpha_a^2$. Here α_p is total parameter, α_b the "reflected parameter" as discussed by Motz, α_s the range straggling parameter, and α_a the angle straggling parameter. The range straggling parameter was derived from the graphical presentation of extrapolated range minus mean range as a function of mean range for integral curves given by Livingston and Bethe. The angle straggling parameter was calculated in the manner indicated by Livingston and Bethe for "good geometry" in blithe diregard of the fact that throughout this work the geometry was in truth "poor". Livingston and Bethe point out the difficulty of obtaining good geometry with heavy nuclei (as heavy as sodium), and point out also that with heavy nuclei the recoil energy is not very significant. The author wishes to point out that the notion of a limiting angle for good geometry, i.e., one for which a maximum penetration of particles of a sharp group exists, depends upon the fact that as the angle of emission is decreased the particles (though their energy increases) traverse the absorbing material more and more obliquely. In the usual experimental arrangement (and in that used here) part of the stopping material is in the aluminum foil windows on the bombardment chamber and on the counter. These foils are both bowed by atmospheric pressure in such a way that particles near the limiting angles of emission

traverse this part of the absorbing medium not only near normally but in a region where it is thinned by stretching. The limiting angle for maximum penetration will also depend in small degree (not easily calculated) upon the shape of the sensitive volume of the counter, and this in turn will depend upon the voltage applied to the counter. For all these reasons it is submitted that the use of the "good geometry" approximation for the angle straggling parameter will not lead to gross error.

The curves marked A'B'C' of Figure 5 are predicted group widths in centimeters for ideal geometry (no angle straggling), for the best geometry used in this work, and for the worst geometry used in this work, respectively. The curves A,B, and C are the same quantities converted to kev by the value of dE/dR given for the same mean range.

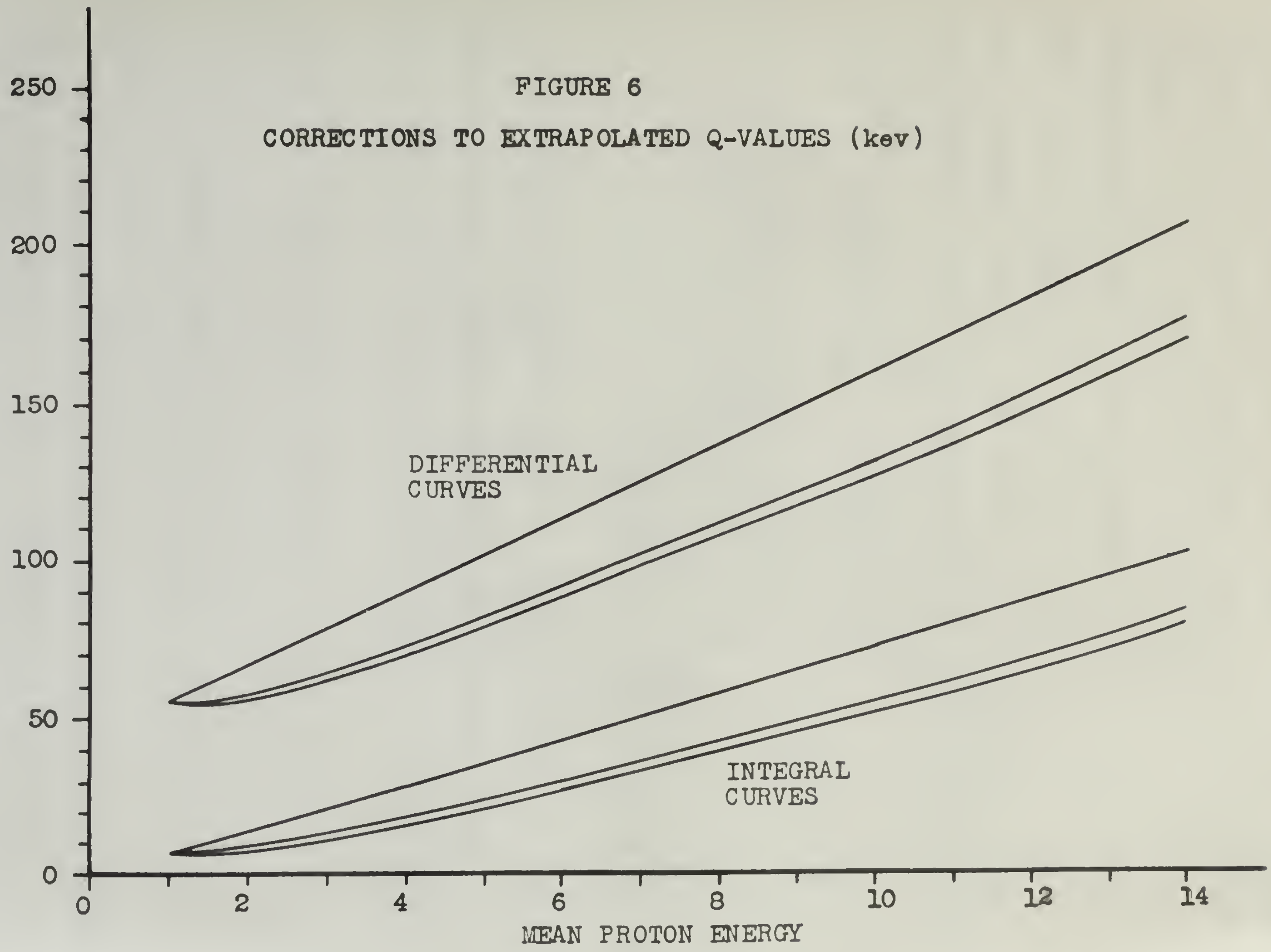
These curves are used to derive the curves of Fig. 6 in the following fashion: One assumes that an integral curve of a proton group has been taken and the mean energy determined from the mean range. This energy is combined with the mean energy of the beam to give the best value of energy release Q for the group. Then for Cl^{35} (d,p) Cl^{36}

$$Q = \frac{37}{36} E_P^{mean} - \frac{34}{36} E_D^{mean}$$

TRA

FIGURE 6

CORRECTIONS TO EXTRAPOLATED Q-VALUES (kev)



using conventional non-relativistic momentum-energy relations and mass numbers for masses. An "extrapolated integral" Q^{ei} may be calculated using the extrapolated energy of the integral curve of the group and the extrapolated energy of the integral curve of the beam: this will be identical with the Q obtained from the mean energies but for the effect of straggling. From the relations for Gauss distributions discussed above

$$\begin{aligned} Q^{ei} &= \frac{37}{36} E_p^{ei} - \frac{34}{36} E_D^{ei} \\ &= \frac{37}{36} E_p^{m} + \frac{37}{36} \cdot \frac{1}{2}\sqrt{\pi}\,\alpha_p \\ &\qquad - \frac{34}{36} E_D^{m} - \frac{34}{36} \cdot \frac{1}{2}\sqrt{\pi}\,\alpha_B \\ &= Q + \frac{37\sqrt{\pi}}{72}\alpha_p - \frac{34\sqrt{\pi}}{72}\alpha_B \end{aligned}$$

where α_p is the proton distribution parameter and α_b is the beam distribution parameter. The difference

$$Q^{ei} - Q = \frac{\sqrt{\pi}}{72}\left(37\alpha_p - 34\alpha_B\right)$$

is the correction which must be applied to the Q^{ei} to obtain the best value of Q.

At the other extreme, for a true differential curve of the proton group, one may calculate in the usual fashion an "extrapolated differential" Q^{ed} from the extrapolated energy of the differential proton curve and the

extrapolated energy again of the integral curve of the beam. The extrapolated energy of the differential curve of the beam is not used because it is not measured directly and would also introduce unnecessary complication.o One now has

$$Q^{ed} = \frac{37}{36} E_p^{ed} - \frac{34}{36} E_D^{ei}$$

$$= \frac{37}{36} E_p^{m} + \frac{37}{36}\sqrt{2}\,\alpha_p - \frac{34}{36} E_D^{m} - \frac{34}{36}\cdot\frac{1}{2}\sqrt{\pi}\,\alpha_B$$

$$= Q + \frac{37}{36}\sqrt{2}\,\alpha_p - \frac{34}{36}\cdot\frac{1}{2}\sqrt{\pi}\,\alpha_B, \text{ and}$$

$$Q^{ed} - Q = \frac{1}{36}\left(37\sqrt{2}\,\alpha_p - 17\sqrt{\pi}\,\alpha_B\right)$$

gives the correction to be applied to Q-values computed in this manner to obtain the best value of Q. For a given mean proton energy, Gauss distribution, and otherwise stable conditions the error in Q calculated from extrapolated proton energy and extrapolated integral beam energy must lie between these extremes. In principle this involves a variation of Δz from 0 to infinity; practically, variation of Δz beyond about 2 has no effect on the extrapolated range. On Fig. 6 are plotted the corrections to be subtracted from the approximate Q - values as functions of mean proton energy. The values of α_p were taken from Figure 5; the value of α_b

used was 90 kev, a mean between that calculated for the free beam and that for the beam when slowed to 3 Mev. Again the three curves in each group are for ideal geometry, the best geometry actually used, and the worst geometry actually used, respectively, in ascending order.

A feature of these plots which strikes the eye at once and can be checked with a straightedge is that they are straight lines within 5 kev over the whole useful range (2 to 14 Mev). The lines for the integral curves converge toward the origin as one would expect; those for the differential curve are nearly parallel to them but converge slightly toward the low energy end because the difference between extrapolated values of integral and differential curves is 0.53 α, as has been shown.

From these considerations a blunt-instrument-type but nonetheless adequate approach to the problem of obtaining accurate Q values is derived. The additional assumption is required that even though the energy distribution of the beam is not a Gaussian it will behave like one when combined with Gauss distributions (i.e., subjected to straggling). This is an assumption of the sort which is often said hopefully to be " not too bad." In suppost of the assumption is offered the fact that the high energy profiles of the measured beam distributions are mostly fairly close to Gaussian (the distributions of Figure 4 are not to be taken as typical; they were

the worst encountered), and it is this part of the distribution which is most strongly influencing the extrapolated value (provided the target is sufficiently thin). The relation

$$X_{ext} = X_o + \frac{\sqrt{\pi}}{2} \alpha$$

for integral curves was found, using values for α measured from constructed differential curves, to be accurate to 5% for the actual beam distributions.

The relation for differential curves

$$X_{ext} = X_o + \sqrt{2}\, \alpha$$

is readily seen to be 10% off for an isosceles triangle and only 40% off for a perfectly square distribution!

The method proposed, then, for analyzing this sort of data is very simple. The chief requirement is to establish one Q-value accurately. More are helpful, but one is enough. If the Q-value is established from the mean range of an integral curve, the extrapolated value may be used to calculated the $Q^{ei} - Q$ for the particular mean energy and also to establish the α_p for the group. From the α_p is obtained Q^{ed} for the same mean energy, using

$$Q^{ed} - Q^{ei} = \frac{37}{36} \cdot 0.53 \alpha_p$$

in the case of Cl^{35} (d,p)Cl^{36}.

A plot similar to Figure 6 may then be made: the $Q^{el} - Q$ will be given by a straight line through the origin and the experimental point. The $Q^{ed} - Q$ line may be drawn parallel to this through the one point already obtained - this will result in a maximum error of 50 kev - or for greater refinement one may compute a second point for a different mean energy using the conventional methods to estimate a new α for the different conditions of straggling. One may also obtain the second point of the $Q^{ed} - Q$ line experimentally if there is a second group sufficiently isolated to permit running of an integral curve.

Any run subsequently made with the same geometry will then, whatever the degree of peaking, have a corresponding line somewhere between the extremes. Measurement of the "extrapolated Q" of a group for which an accurate Q value is available will locate one point of the line. If Δz were constant over a given run, the correction line for the run would be the locus of points lying above the lowere limit by a constant fraction of the distance between the limits. Values of this fraction as a function of Δz are given by the plot of "z_{ext} relative" of Figure 3. Unfortunately it is not Δz which remains constant over a run but ΔR, the counting interval in range, which determines Δz by the relation

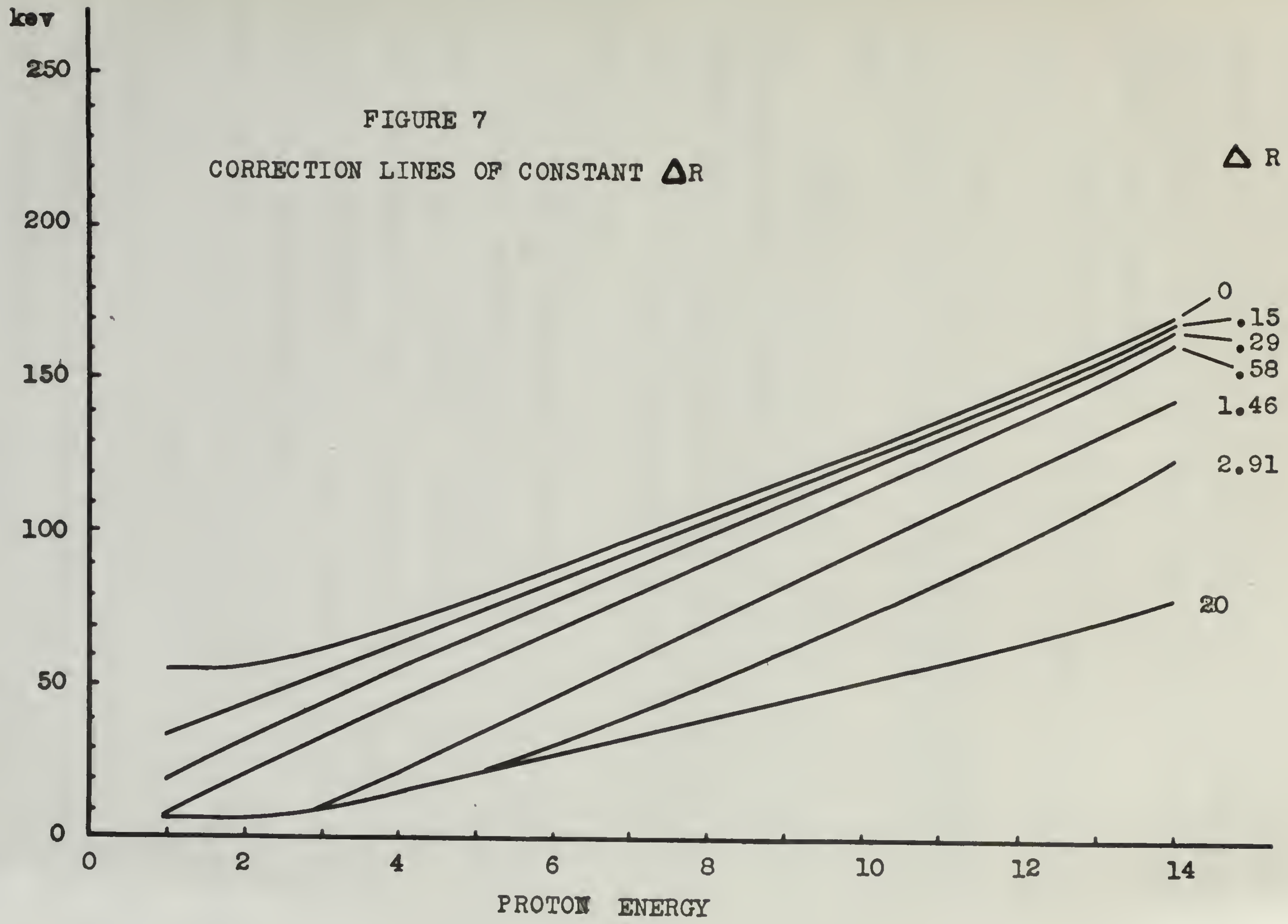
$$\Delta Z = \frac{\Delta R}{\alpha}$$

where α is the group parameter in centimeters. It is this effect which is responsible for the diminishing resolution atsshort ranges (along with increasing dE/dR and in spite of decreasing straggling), at least partly for the increased yields at short ranges, and for the fact that the structure of the beam seems to show up only in long range groups. Correction curves for lines of constant ΔR for the ideal geometry case of Figure 6 are shown on Figure 7. Here the bluntness of the blunt instrument becomes apparent: these are very close to straight lines also, and all the important ones pass within 25 kev of the origin. The second and crucial step of the method, then, and one which can be accomplished without completing the first, is this: Through a point on a correction line established from an accurate Q and an "extrapolated Q" for a given run, draw a straight line through the origin. Corrections to all extrapolated Q values for the run will then be read directly from the line as functions of the proton energy.

If the instrument seem painfully blunt to the reader, let him remember the shapesof the beam distribution and its dubious relation to the shape of the Gauss distribution for which this analysis was reasonably rigorous up to the preceding paragraph. What has been established is that for extrapolated Q values determined by the methods used in this laboratory, but with a beam of deuterons whose numbers-energy distribution is describable by a

FIGURE 7

CORRECTION LINES OF CONSTANT ΔR



Gauss function, corrections can be calculated and shown to tend toward zero for small proton energies. As the actual beam distribution is indescribable and varies in time besides, but has in common with the Gauss distribution the attributes of

1. Being in effect bounded by an upper and lower limit

2. Rising from the limits to maximum intensity at and around a mean (actually, by our definition, median) value

3. Having a width or parameter which is related to the extrapolated intercept of the integral curve in a regular way; the existence of such corrections and such tendency are inferred for actual Q-values measured in this laboratory.

The several objections to the method and manner of the inference would be more serious if the method laid claim to greater precision. One is the impropriety of using the root-sum-square combination (K2) of parameters of superposed distributions one of which is patently not of the same kind as the others. A second is the impropriety of using this method of combination of distributions one of which (that of the beam), even if it were really "closely Gaussian", is not generated by the random statistical processes assumed in the development of probability theory. These are valid objections, and because of them the claims of precision of the analysis and indeed of the

ultimate precision of the observations with the apparatus are limited.

Certain other corrections have been calculated; these are of less importance than the change of group shape and range by peaking discussed above.

The error in calculated Q-values introduced by neglecting relativistic corrections has been calculated for 4 Mev deuterons to be 1.7 kev for 10 Mev protons from $Cl^{35}(d,p)Cl^{36}$ and 2.6 kev for 12 Mev alpha-particles from $Cl^{35}(d,\alpha)\,S^{33}$.

A correction for poor geometry has been estimated also. As Bethe has shown, poor geometry is such that particles of an angle of emission corresponding to a maximum penetration into the detector can reach the detector. As Bethe has also shown the effect is to create a rectangular distribution from a group of particles originally sharp, the upper limit of which distribution is the true range of the group and the width of which is

$$R_o\,(1-\cos\chi_o)$$

if one neglects the dependence of energy on the angle χ made with the axis of the detector by the particles. Here R_o is the "true range" of the particles and χ_o the limiting angle of detection. The dependence of energy on direction changes the maximum range from R_o to $R_o(1-\frac{1}{2}\cos^2\theta_o)$ where θ is the angle corresponding to maximum penetration. The total effect then is to generate a rectangular distribution whose mean range is

$$R_o\left[1-\tfrac{1}{2}\cos^2\theta_o - \tfrac{1}{2}(1-\cos\chi_o)\right]$$

When combined with the large distributions already discussed this has the same effect as an increase of counting interval on the shape of the observed curve but will shift its whole position by an amount corresponding to

$$\frac{1}{2}\cos^2\theta_0 - \frac{1}{2}(1-\cos\chi_0)$$

This is a correction which does not go to zero with zero energy and which must be applied to the ranges of integral curves used for establishing accurate Q-values as well as to others. It is however small, averaging about 1 centimeter for the worst geometry used in this work and being of the order of 1 or 2 millimeters for the best.

Target thickness corrections have been discussed by Motz with respect to this kind of work. The effect of small target thickness has been shown to be equivalent to that of a small increase in ΔR on the observed shape of a group; this may be considered as a change in distribution parameter as suggested by Motz or allowed to be absorbed by the method of determining the approximate correction. As target thickness in this work was never over 100 kev and was usually much less, the correction for it was considered to be included in the overall correction.

The matter of counter depth has been discussed at great length by several writers (H1, H2, M2). It was not deemed profitable to make precise determinations of the variations of this quantity as the correction involved

is very small. The magnitude of the counter depth for alpha particles was calculated by comparison of the N^{14} (d,α) C^{12} end group recently published by Malm and Buechner. (M6. Also see M4 for others). A mean value for protons was estimated and used for all the work.

A final correction of small magnitude has been applied to Q-values for groups assigned to Cl^{37}. As the masses of the two isotopes of chlorine are so close together percentagewise, purely as a matter of convenience all Q- values were originally calculated as though for Cl^{35}. After assignment of the groups the small corrections were made (amounting to as much as 14 kev) to those assigned to Cl^{37}.

It is believed that Q-values determined by this method should be accurate to within 80kev over the whole range of values measured. The estimate is based upon consideration of all the sources of error which can be anticipated. Errors in the excitations of the excited states will be comparable, with some reduction because the excitations are determined from differences of Q-values and the effects of systematic errors will tend to be compensated.

2. Experimental Results

A total of 18 proton groups were observed. Figure 8 shows a numbers-vs.-range curve for one of the enriched AgCl targets. Figure 9 shows a similar curve for a gas target whose chlorine content was 87% light isotope. The solid target curve shows how the closely spaced groups were resolved by constructing symmetrical groups from right to left. The gas target curve shows the vertical axes of symmetry of the constructed groups. This particular curve was chosen to illustrate the gas target data because it was one of the few showing a single run covering the whole range of proton energy. It shows only medium resolution, as it was made with a thick target (30 kev) and less-than-average peaking. The difficulties of resolution were not much reduced even in the best runs, however. Q-values for the groups observed were (all calculated as though from Cl^{35} in Mev:

A.	6.27 ± .09	K.	2.57 ± .09
B.	5.51 ± .08	L.	2.39 ± .09
C.	5.09 ± .08	M.	1.88 ± .08
D.	4.72 ± .08	N.	1.45 ± .11
E.	4.30 ± .09	O.	1.11 ± .08
F.	3.79 ± .09	P.	0.78 ± .09
G.	3.42 ± .09	Q.	0.31 ± .1
H.	3.01 ± .09	R.	0.07 ± .09
J.	2.71 ± .11	S.	-0.36 ± .1

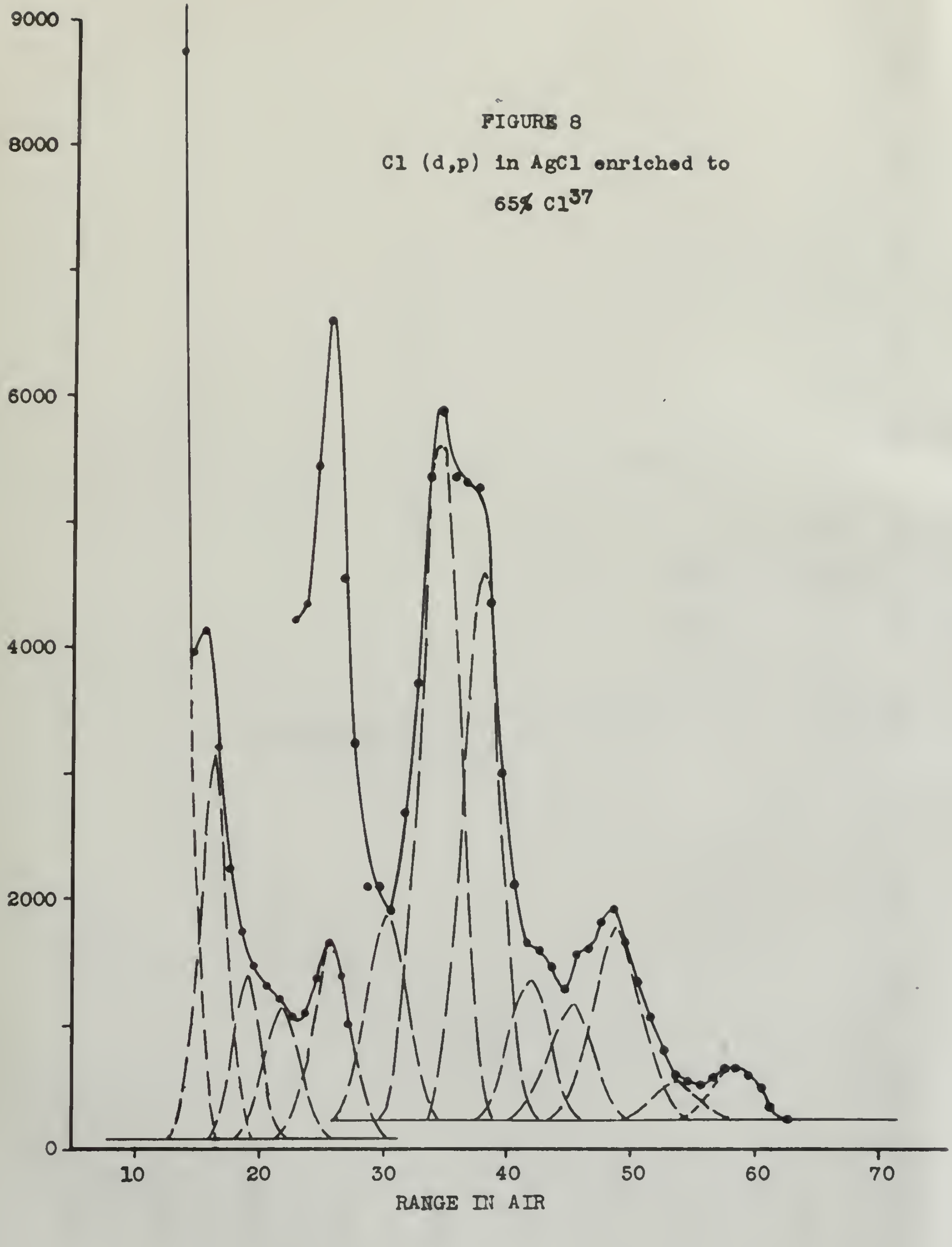


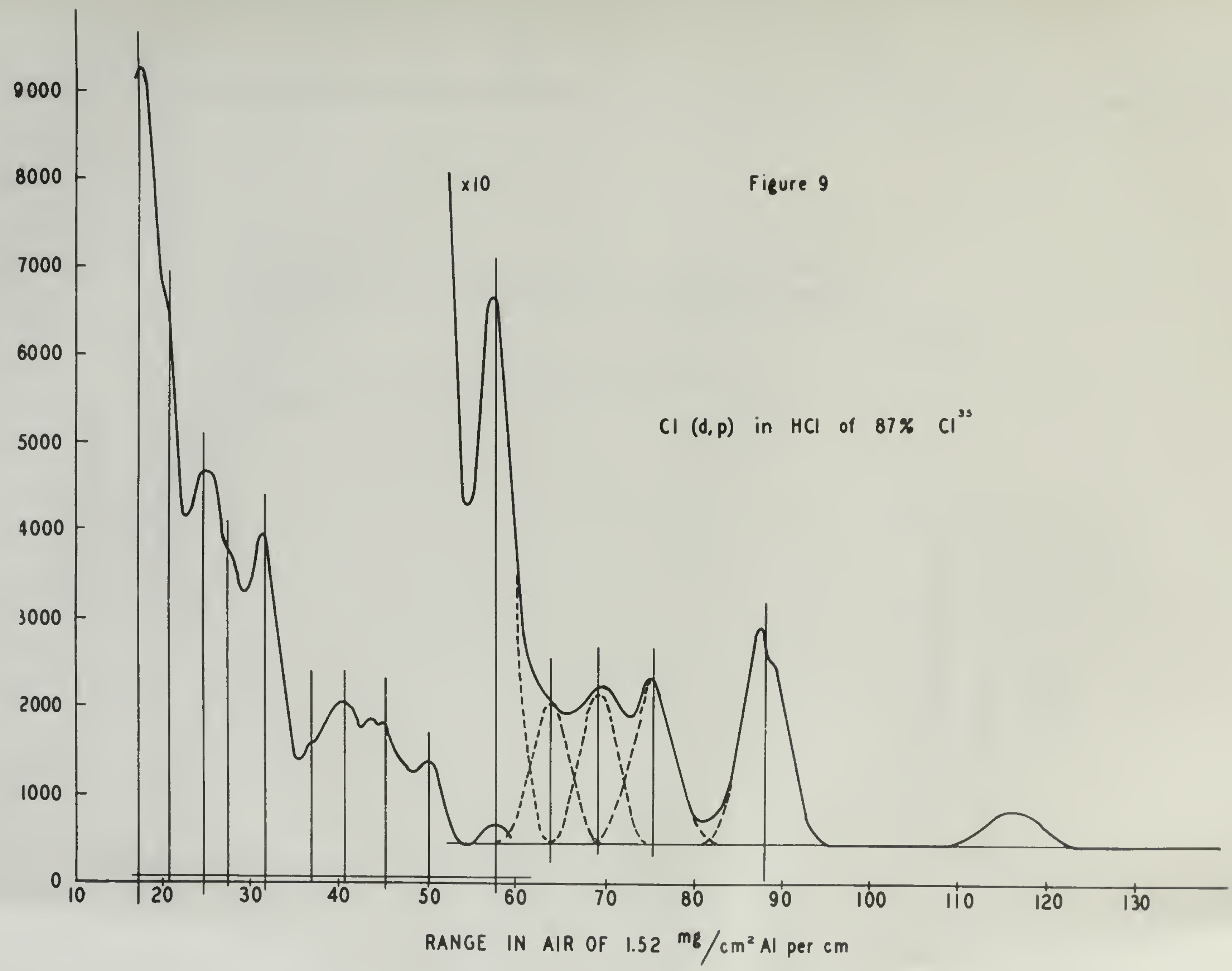
FIGURE 8

Cl (d,p) in AgCl enriched to

65% Cl^{37}

Figure 9

Cl (d, p) in HCl of 87% Cl^{35}



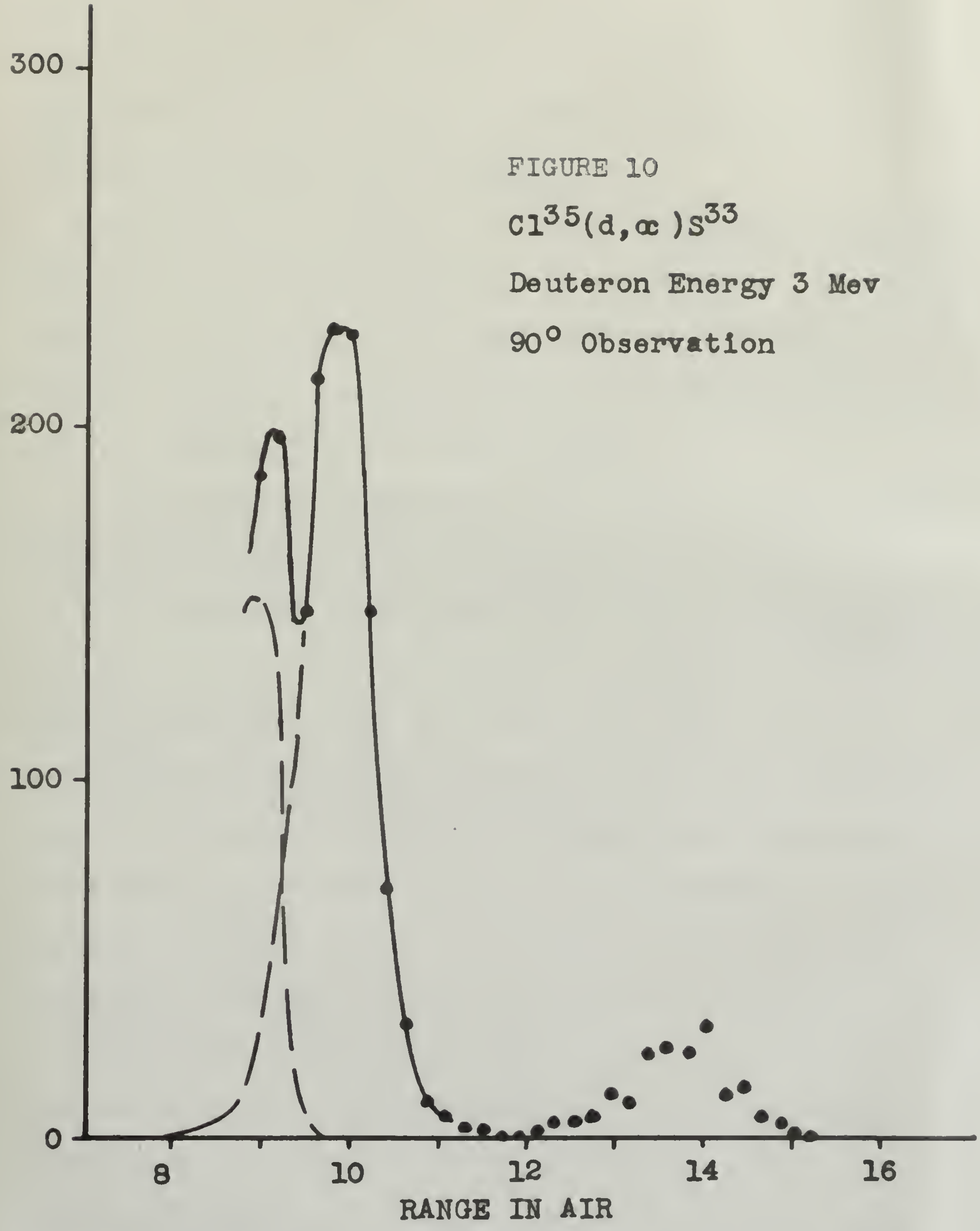


FIGURE 10

$Cl^{35}(d,\alpha)S^{33}$

Deuteron Energy 3 Mev

90° Observation

The errors listed reflect standard deviations in the observed values as well as the errors and uncertainties previously estimated. Groups Q and S were observed in just one run: the only one on enriched silver chloride to extend to such low energies.

Analysis of the groups to effect assignment to isotopes has proved difficult and not very profitable. The information available from the solid silver chloride was limited (3 runs) in quantity and also in dependability by the decomposition of the targets.

The nearly simultaneous final failure of the cyclotron and failure of the column which was enriching HCl in Cl^{37} effectively prevented obtaining any more heavy isotope data, either with locally prepared targets or with HCl prepared from the Oak Ridge AgCl. The data available were therefore subjected to as searching analysis as possible in order to extract what information there was. The method adopted was to construct a large matrix-like array whose element R_{ij} was the ratio of the yield of group "i" to that of group "j" in the region above the diagonal, where were entered these ratios for runs on targets containing a preponderance of Cl^{35}; and whose element R_{ji} was the same ratio in the region below the diagonal, where were entered the ratios for runs on the enriched AgCl. This arrangement permitted direct comparison of relative yields for all pairs of groups for all runs. This comparison was

supplemented by visual comparison of the curves for the runs themselves.

The maximum ratio of relative isotopic concentrations in the different targets was about 12.5 (from 87:13 to 35:65), a variation which should in principle allow easy discrimination of groups from the two isotopes. The effect, already mentioned, of the vagaries of the method of resolving the closely spaced groups upon the relative yields of the groups was such as to cause the apparent relative yields to vary by a factor of as much as 2 from run to run. In no case was a variation of as much as 12.5 observed, even between the extremes of isotopic concentrations, so the assignments to isotopes of the groups and consequently of the excited states can be regarded at present as no more than "educated guesses". The assignments as made, together with relative yields and excitations, are given in Tables II and III.

Table II Cl^{35} (d,p) Cl^{36}

Q		Relative yield	Excitation	(B6)level	Shrader
A	6.27	16	0	0	0
B	5.51	8	0.76	0.79	0.96
C	5.09	7	1.16	1.14	
D	4.72	6	1.55	1.63	
E	4.30	20	1.97	1.94	
F	3.79	50	2.48	2.44	
G	3.42	65	2.85	2.84	
H	3.01	80	3.26	3.05	
J	2.71	55	3.56	3.55	

Table II, cont'd.

Q		Relative yield	Excitation	(B6)level	Shrader
O	1.11	165	5.16	(4.10)	
P	0.78	175	5.49	(4.30)	
R	0.07	335	6.20	(4.94)	

The agreement with the Chalk River groups is seen to be good with the exception of groups D and H; also no counterparts to the last 3 gamma-rays observed by that group were found.

Table III Cl^{37} (d,p) Cl^{38}

Q		Relative yield	Excitation	Shrader
F	3.77	10	0	0
K	2.55	23	1.22	1.00
L	2.37	97	1.40	
M	1.86	107	1.91	1.92
N	1.43	32	2.34	
Q	0.29	120	3.48	
S	-0.38	900	4.15	

Group F is shown as belonging to both isotopes because it is believed to be a very close doublet from the change of shape shown by it in going from Cl^{35}- rich targets to Cl^{37} rich targets. This assignment is also the only one which could be made consistent with existing mass data. That it agrees very closely with these data is probably fortuitous, because of the inaccuracy introduced by the tacit assumption that both elements have the same energy in a doublet which is wide enough to

change its shape appreciably with a change in relative magnitude of the elements.

The reaction Cl^{35} (d,α) S^{33} has also been observed down to 8cm range. Two groups were found, having Q-values of 8.47 ± .1 Mev and 7.84 Mev. No error is assigned to the 7.84 Mev group as it is considered probable that its shape was distorted by the N^{14} (d, α) C^{12} group with Q = 9.14 Mev. When combined with the 8.47 value for the ground state it gives 0.63 Mev for the first excited state, in reasonable agreement with Davison's value of 0.79 (D2). The Q-value of 8.47 Mev for the ground state was established by direct comparison with the N^{14}(d,α) C^{12} ground state group with an accurately established Q of 13.58 Mev (M6).

Some mass data are also available from the three ground state Q-values reported here. Values for the basic masses used,

n^1 1MU + 8.98 mmu
H^1 1 + 8.14 mmu
D^2 2 + 14.72 mmu
He^4 4 + 3.91 mmu,

are those of Tollestrup, Fowler, and Lauritsen (T3).

From these the reaction

Cl^{35} (d,p) Cl^{36} Q = 6.74 mmu

yields a mass difference

Cl^{36} - Cl^{35} = 0.99984 mmu

This may be compared with values of

0.99981 (from Shrader and Pollard)
1.00017 (microwave determination, L7)
0.99979 (n, γ from Chalk River group).

Measurement of the Cl^{36} positron (if there were such a positron) would allow checking of mass differences around the cycle $S^{36} - S^{34}$ (L6), $S^{34} - S^{33}$ (D2, L7), and $Cl^{35} - S^{33}$ (S1 and this paper). Measurement of the mass difference $A^{36} - A^{35}$ would also create a closed cycle with $Cl^{36} - Cl^{35}$: $A^{35} - Cl^{35}$ (E1, W4) and $A^{36} - Cl^{36}$ (F2, W3).

The reaction

$Cl^{37}(d,p)\,Cl^{38}$ $\quad Q = 4.05$ mmu

yields a mass difference

$Cl^{38} - Cl^{37} = 1.00251$

which may be compared directly with 1.00228 (from Shrader and Pollard). Better agreement is obtained from the cycle

$$\begin{array}{lll}
Cl^{37} - Cl^{35} & 1.99751 & (T2, G1) \\
A^{38} - Cl^{35} & 2.99443 & (P3) \\
Cl^{38} - A^{38} & .00553 & (H6) \\
\hline
Cl^{38} - Cl^{37} & 1.00250 &
\end{array}$$

The reaction

$Cl^{35}\,(d,\alpha)\,S^{33}$ $\quad Q = 9.10$ mmu

yields the mass difference

$Cl^{35} - S^{33}$ $\quad 1.99829$

This does not agree very well with Shrader and Pollard's value of 1.99896 but may also be compared by combining the two mass differences

$$\begin{array}{lll} S^{33} - S^{32} & 0.99962 & (D2) \\ Cl^{35} - S^{32} & 2.99802 & (E9) \\ \hline Cl^{35} - S^{33} & 1.99840 & \end{array}$$

showing agreement to 0.1 Mev which is within the limits of experimental error.

The effects of impurities on the groups have been evaluated upon the basis of published data on the proton yield from nitrogen and oxygen and from air (D2). Normal nitrogen yields from gas targets purified by freezing appear on Figures 9 and 10. In the case of protons a nitrogen concentration corresponding to a yield of about twice that of the chlorine end group would be required for air contamination seriously to affect the groups. For the alpha-particle yield it was estimated as has already been implied that the nitrogen content was sufficient to displace the low-energy group. The presence of an undetermined but probably small smount of carbon in the gas targets was indicated by

(a) Formation of a yellowish, acetone soluble deposit with an unidentifiable spicy odor on the bottom of the chamber under the beam path

(b) Presence of a distinct pink tinge (going in one instance to a deep cherry red) in HCl frozen after long bombardment. This appeared after the HCl froze and on warming disappeared beforethe HCl sublimed - indicating that the presumed organic chlorine compound could not be removed by pumping on frozen HCl. This unidentified contaminant may have been picked up from stopcock grease or from rubber tubing used in the system.

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